

MAUI

"Modeling, Analysis, and Ultrafast Imaging"

Tom Peterka

Mathematics and Computer Science Division

Argonne National Laboratory

Integrated Imaging Institute Seminar

3/21/16

Team

Modeling

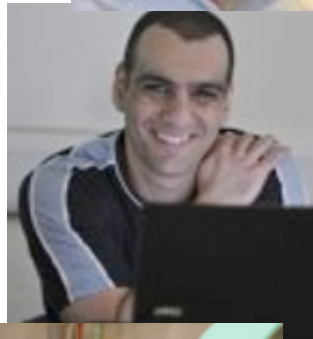
- Subramanian Sankaranarayanan (CNM)
- Kiran Sasikumar (CNM)

Analysis

- Todd Munson (MCS)
- Sven Leyffer (MCS)
- Nicola Ferrier (MCS)
- Youssef Nashed (MCS)
- Tom Peterka (MCS)

Ultrafast Imaging

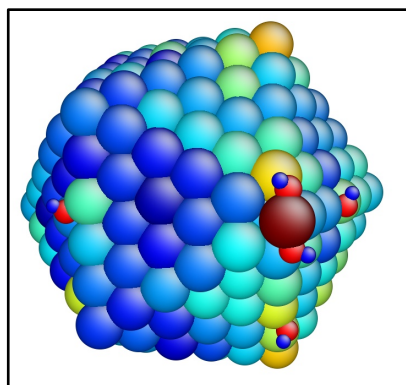
- Ian McNulty (NST)
- Ross Harder (XSD)
- Haidan Wen (XSD)
- Mathew Cherukara (XSD)
- Andrew Ulvestad (MSD)



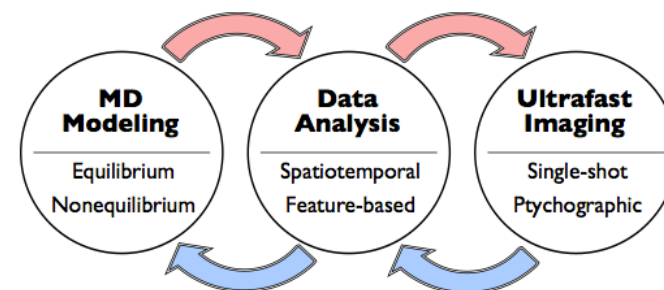
Objectives



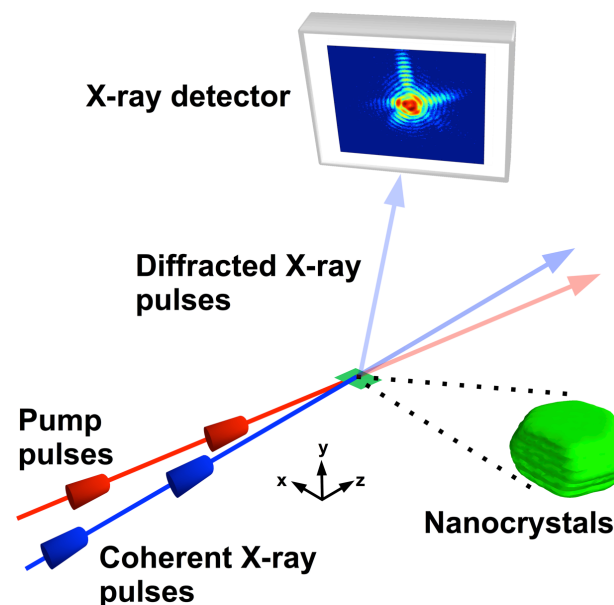
- Pump-probe experiments (Sector 7)
- Reactive MD (LAMMPS Reax)
- Understanding phonon breathing modes
- Understanding reaction-induced strain
- Lattice fitting
- Fast phase reconstruction
- Comparing results in different time/length scales
- Workflow development
- Attracting new talent
- Learning a new language
- Attracting new funding



Displacement of Au atoms after 20 ps in 0.55 M ascorbic acid.



Simulation and experiment are integrated through data analysis.



**J. N. Clark, et al.
Science 341 (6141): 56–59.**

Phonons: Sound Waves that Carry Heat in Crystal Lattice Vibration

- Heat dissipation of next-generation semiconductors
- Conversion of wasted heat into electricity in thermoelectric materials
- Electrochemical processes across liquid-solid interfaces in water
 - phase transitions
 - bond softening/hardening
 - ferroelectricity
 - solid/liquid interfaces
 - heat dissipation
 - phononic local structure
 - phase front propagation
 - spectrometry



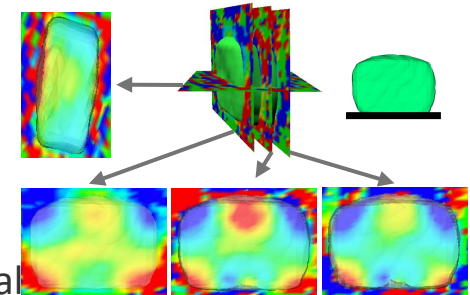
<http://water.org/water-crisis/water-facts/water/>



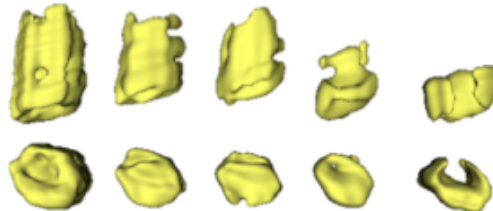
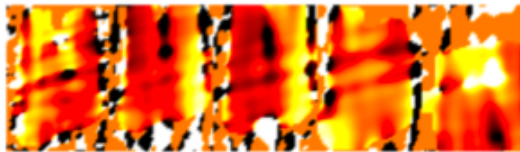
Coherent Diffraction Imaging: Applications

In-situ and operando characterization of materials evolution

- Mechanical response at nanometer length scales
 - Structural response to chemical reactions & in-situ catalysis
 - Decomposition of semiconductors in contact with noble metals
 - Structural changes in crystals due to defect formation and removal
 - Alloying and dealloying
 - High temperature and pressure
- Domain wall (magnetic, orbital, charge) structure in the complex oxides and multi-layer
- Phase transitions vs temperature and magnetic field



Cha, Wonsuk, et. al 2013
Nat Mater 12 (8) 729–34.



Coherent diffractive imaging of solid state reactions in zinc oxide crystals.

Leake, S. J., Harder, R., & Robinson, I. K.

(2011) *New Journal of Physics*, 13(11), 113009

Watari, M., Harder R., et al. (2011).
Nat Mater, 10(11), 862–866.

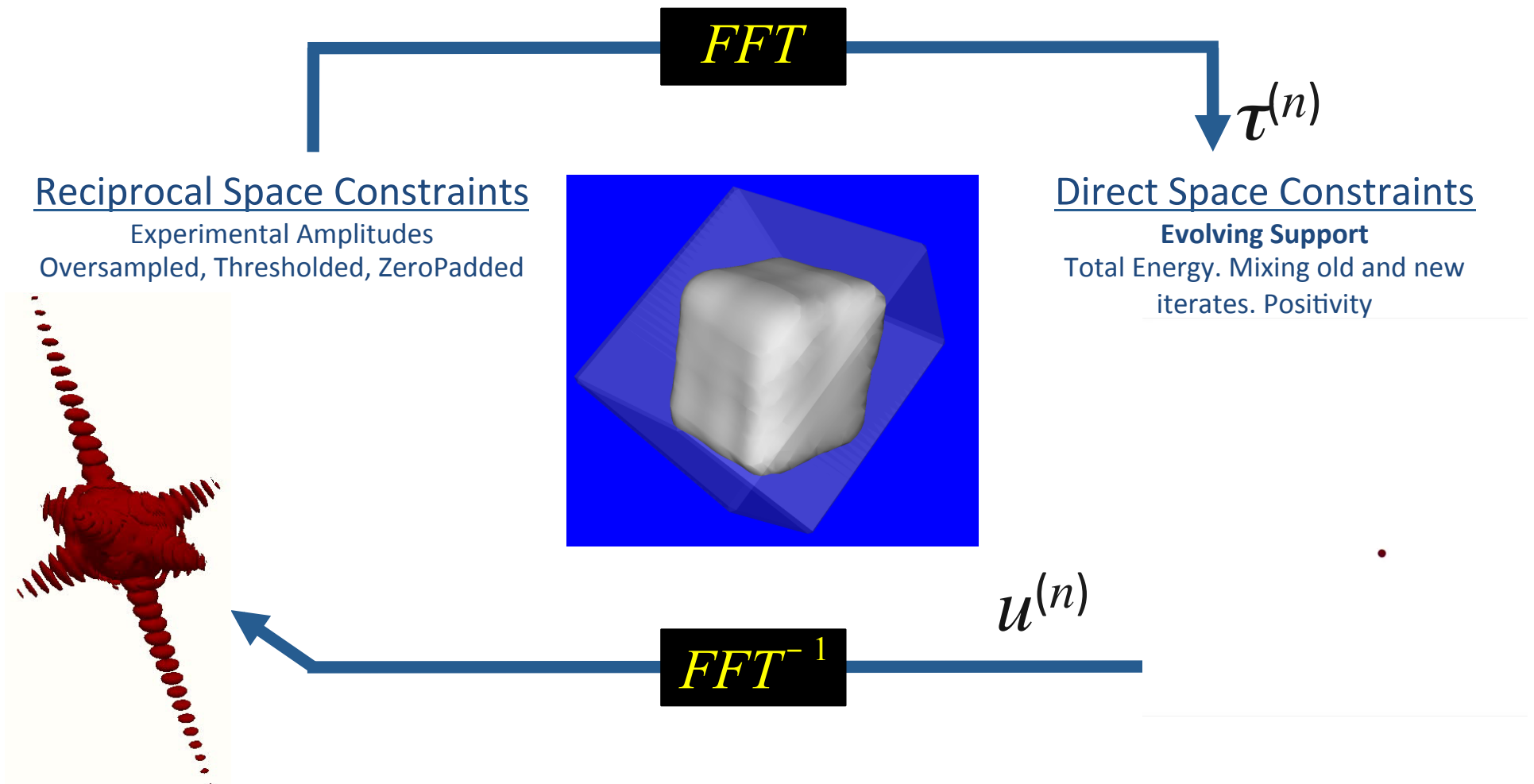
chain thiols, the large stress cannot arise from the van der Waals chain-chain interactions or other weak forces alone, but at least ionic or covalent rearrangements. Indeed the Au-S bond plays a crucial role in SAM formation: the structure of the (1.6 nm) thiolated nanocrystals has its Au-Au spacing disrupted and sulphur intermixed with gold in the outer layer. Our findings support this model and show strong thiol-induced deformations of our 300 nm crystals with strains penetrating more than 20 nm from the outer surface towards the crystal core. The tight-radius spherical parts of the nanocrystals might also undergo strong Au-S intermixing. Our observation of relative contraction of the curved surface regions of Au nanocrystals would indeed be able to provide sufficient stress. reactions involving atomic diffusion of Au at room temperature would also be an attractive explanation of the relatively large expansion seen in both our X-ray and the cantilever experiments.

Our observation of relative contraction of the curved surface regions of Au nanocrystals, illustrated in Fig. 3e, leads to the conclusion that the

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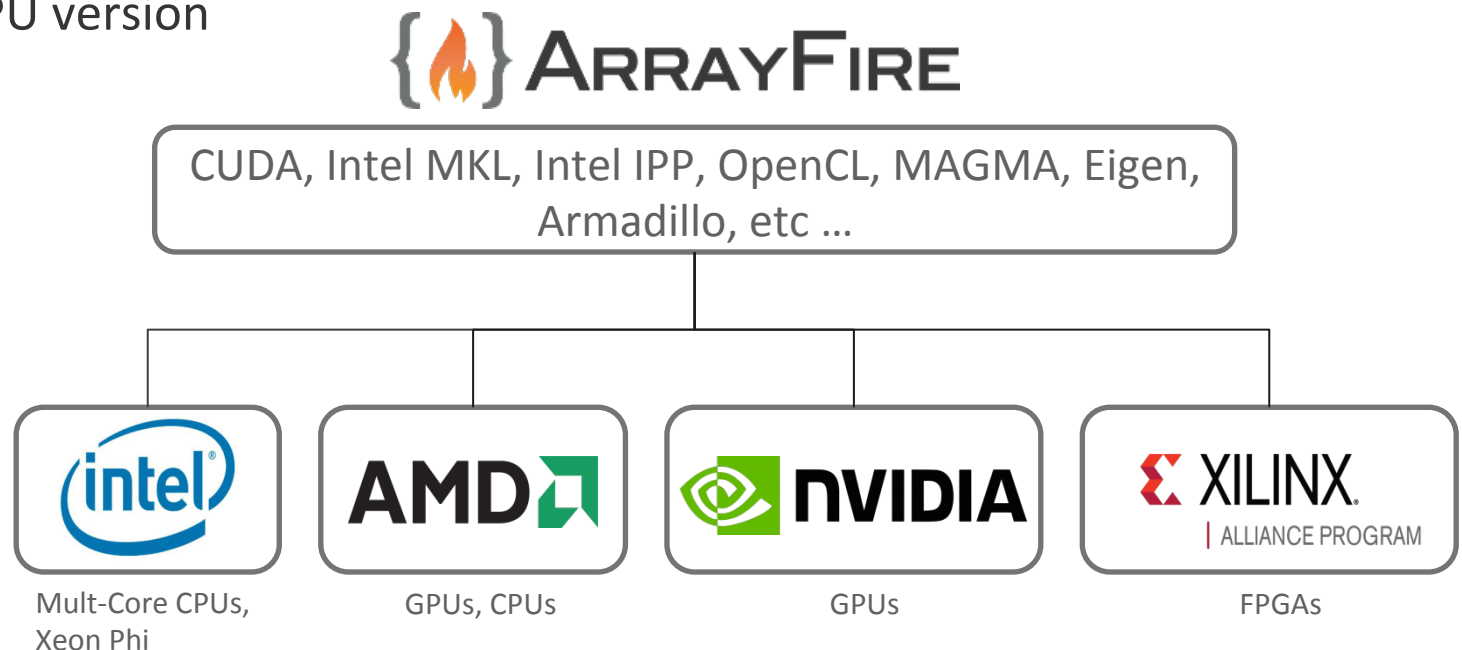
Phase Retrieval: Challenging and Costly Inverse Problem



1000's of iterations and multiple random starts required for high fidelity data!

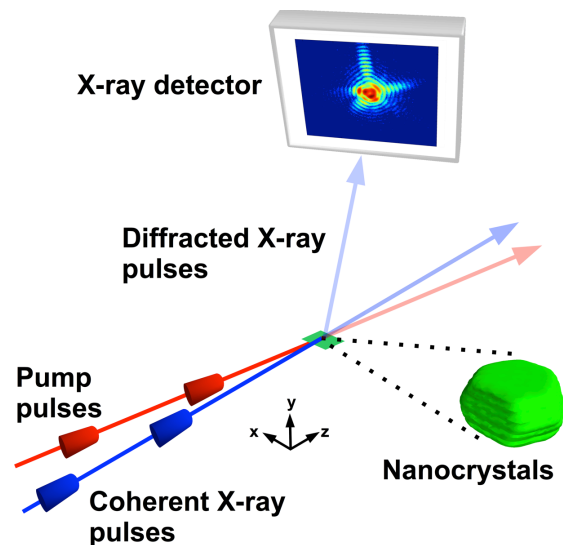
Fast Phasing Library (FPL)

- Group phase retrieval methods and tricks in one library
- Runs on various platforms (from user laptops to HPC systems)
- Hardware-independent code through ArrayFire
- Easy to customize, maintain and extend
- Currently over 100x faster than GPU accelerated MATLAB code, ~500X over the CPU version

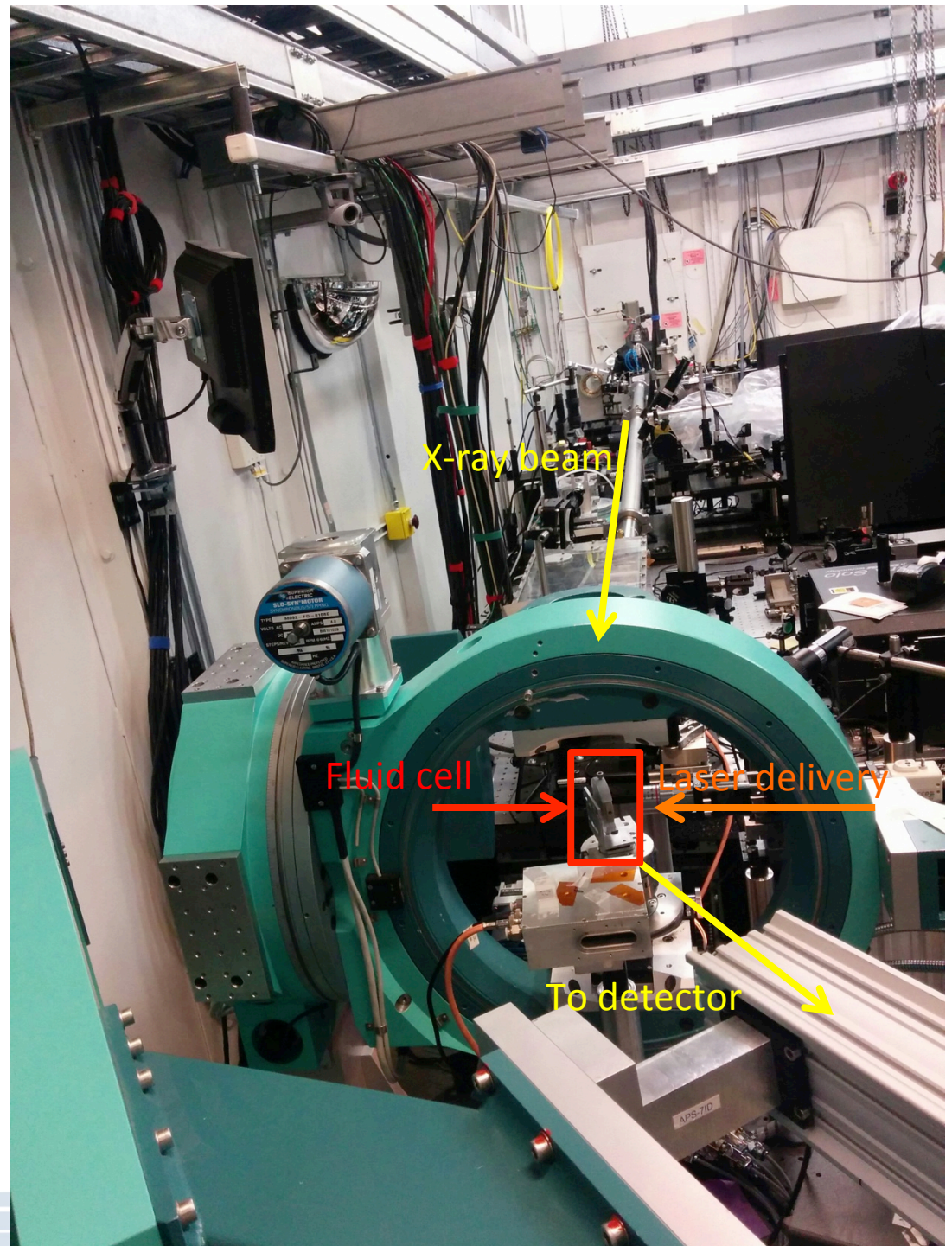


Time-resolved CDI

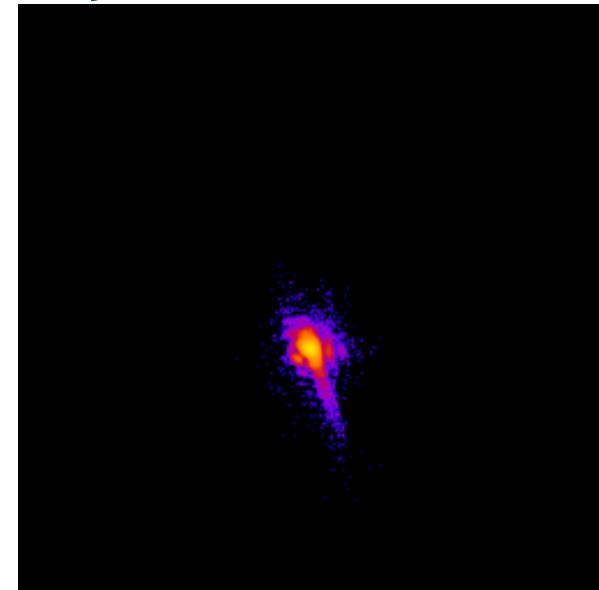
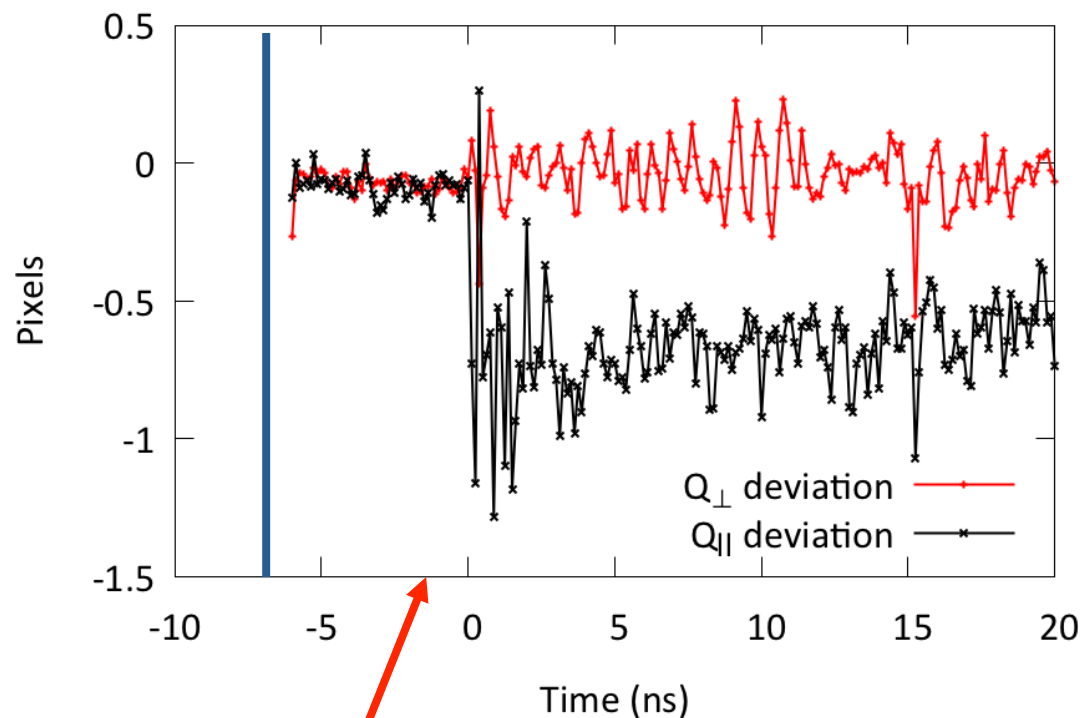
- Laser pump, X-ray probe at Sector 7
- Combine with CDI
- Provide dynamical map of excited modes in sample.
- <10 nm spatial resolution possible with 100 ps temporal resolution.



J. N. Clark, et al.
Science 341 (6141): 56–59.



Case Study 1: Dynamic Response of Au Crystals

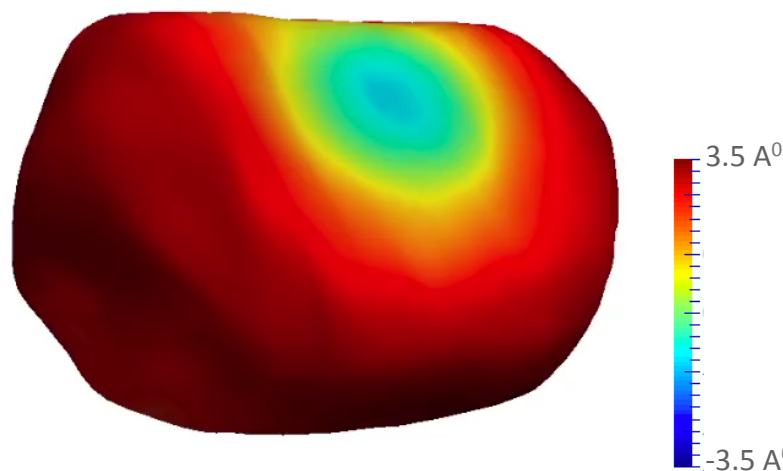


Reciprocal space view

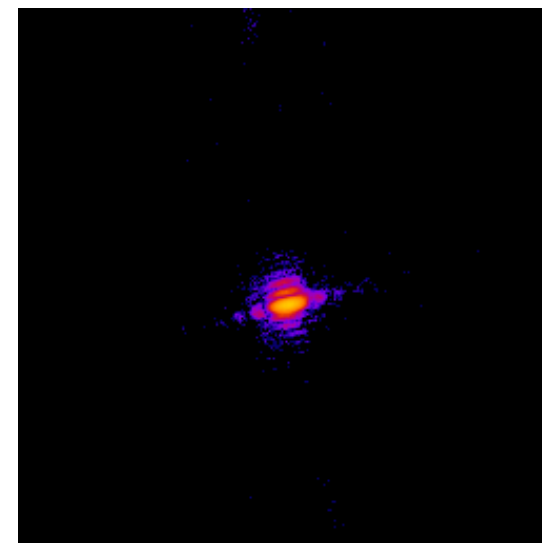
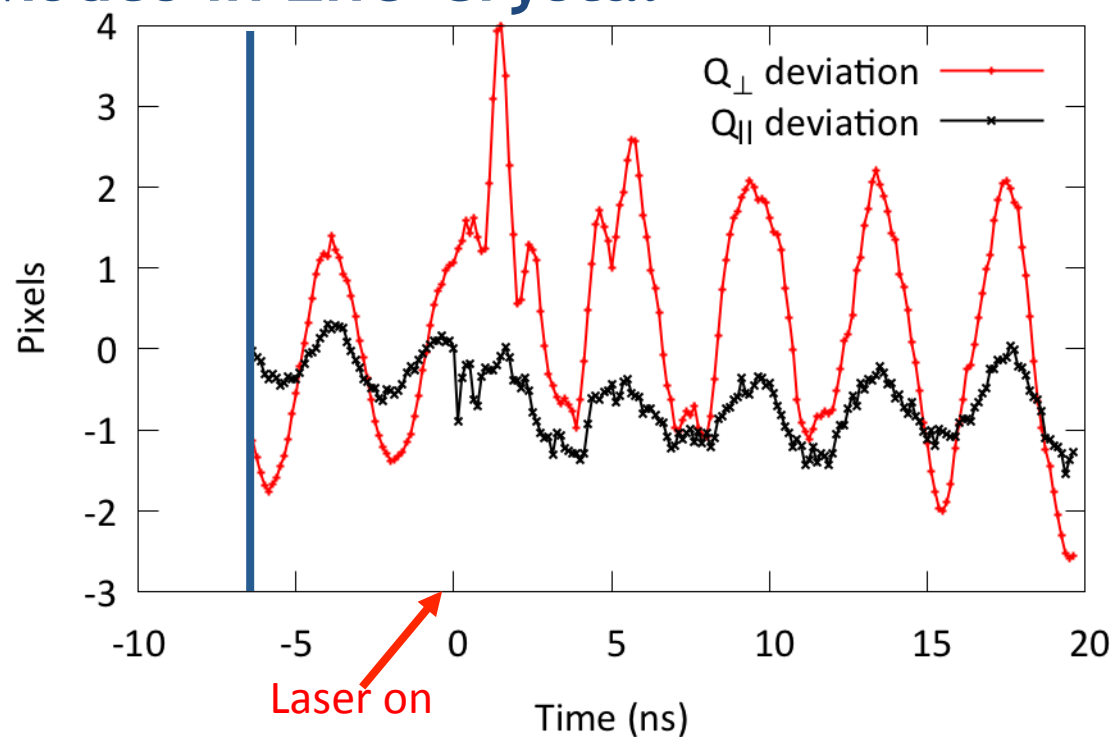
Phase retrieval
at select times

Need to measure the
complete rocking curve at
those points

Real space view



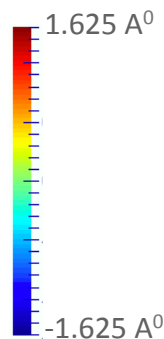
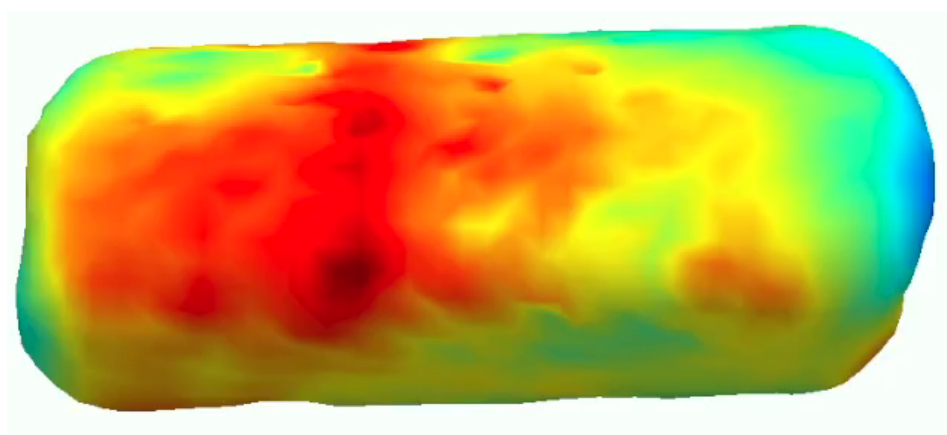
Case Study 2: Volumetric and Deviatoric Deformation Modes in ZnO Crystal



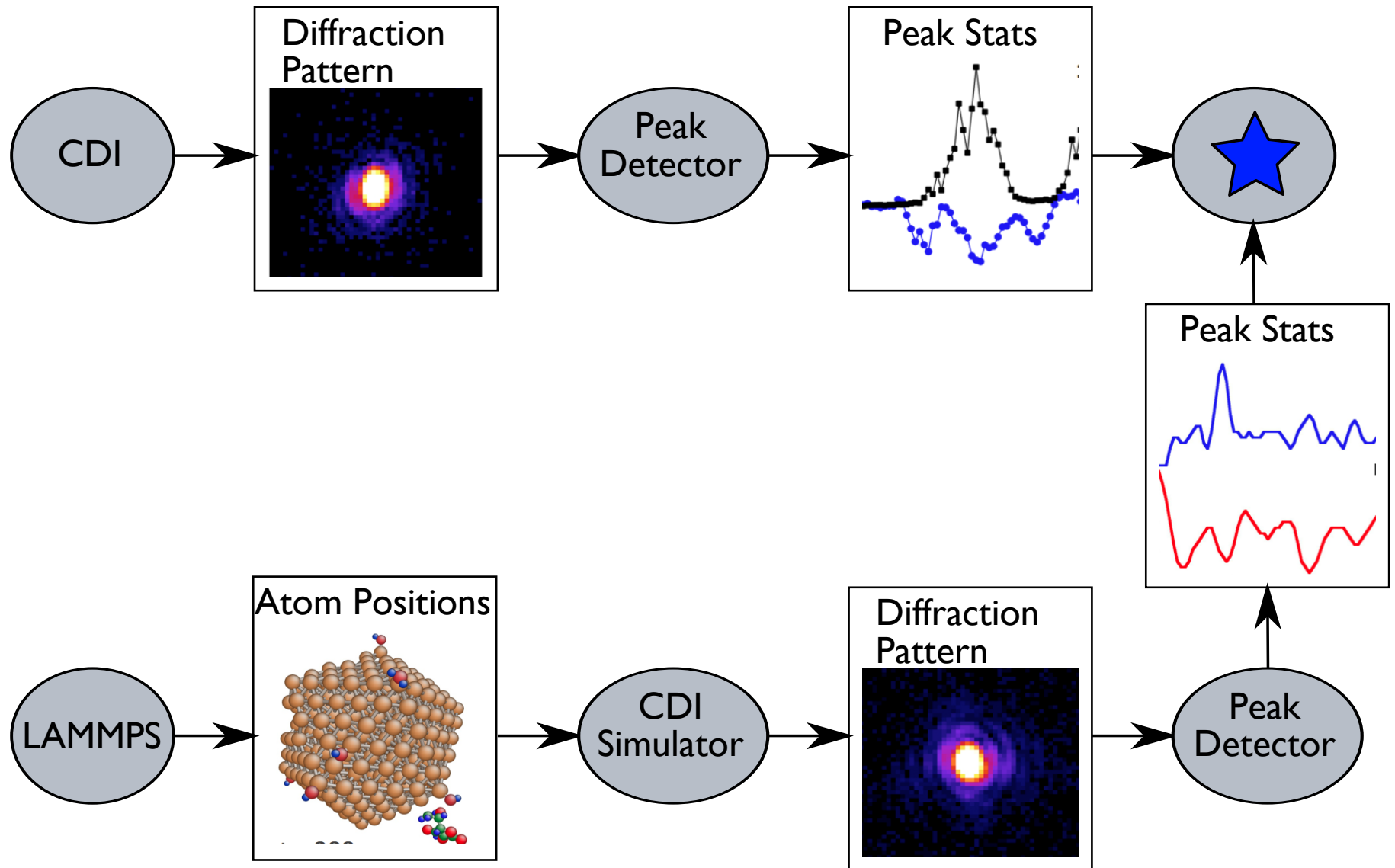
Reciprocal space view

Phase retrieval
at select times

Need to measure the
complete rocking curve at
those points

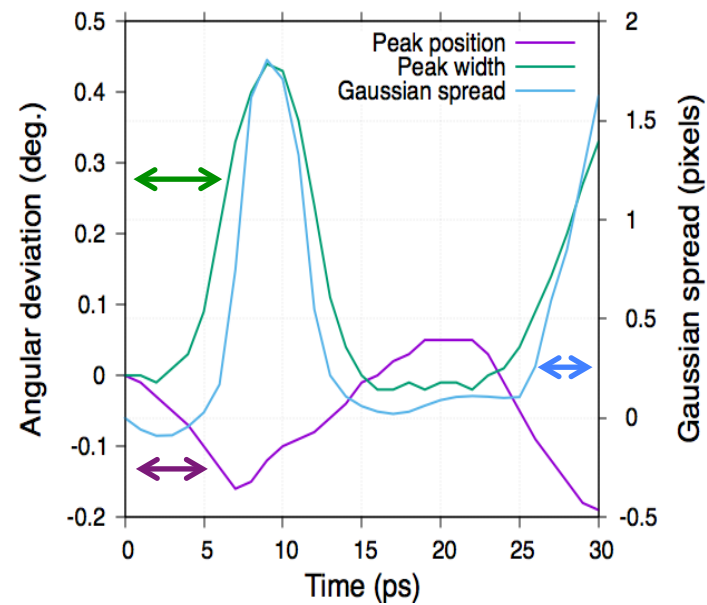
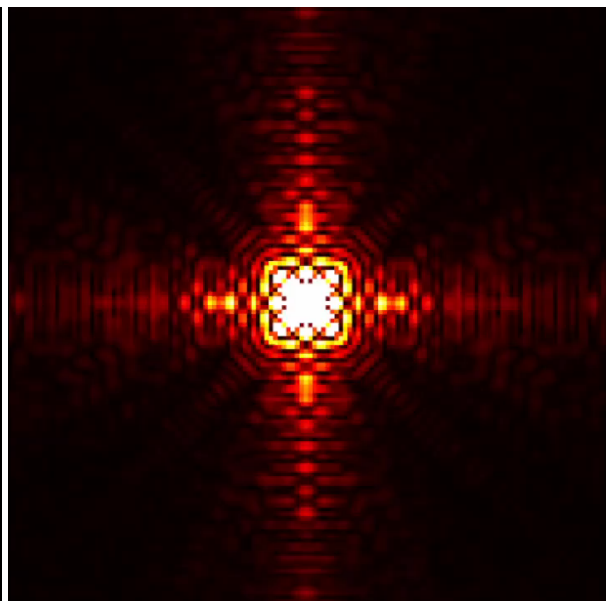
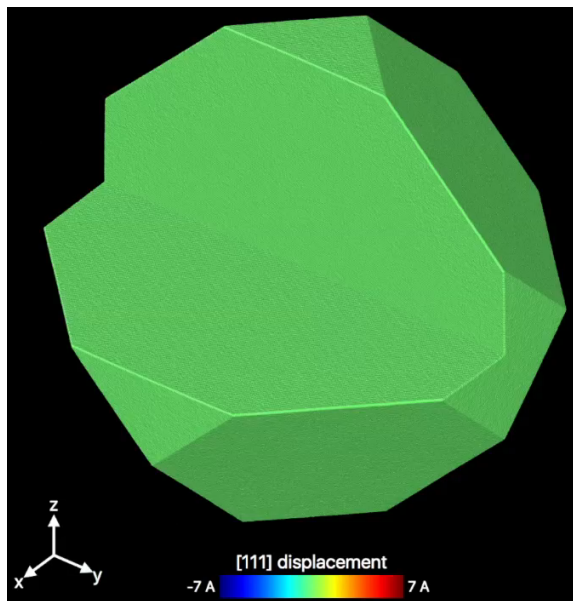
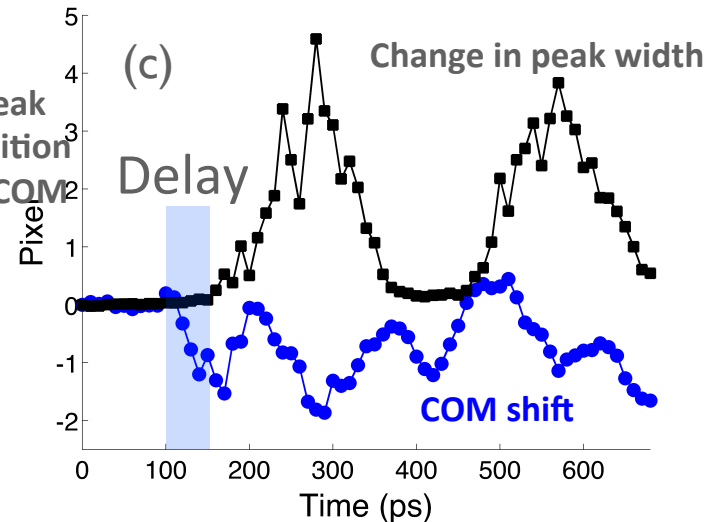
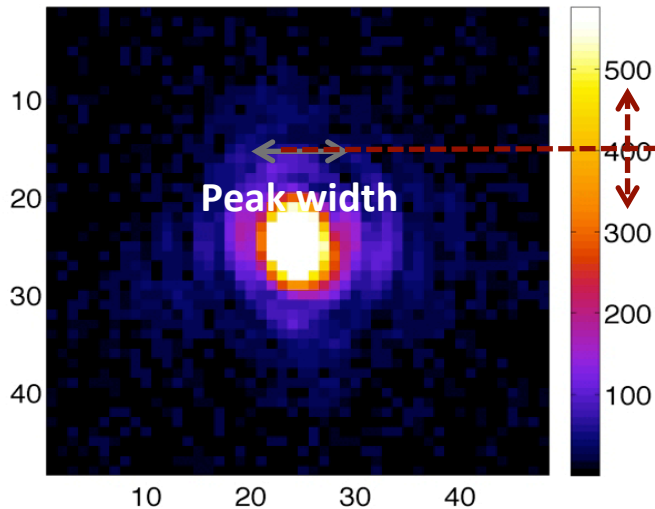
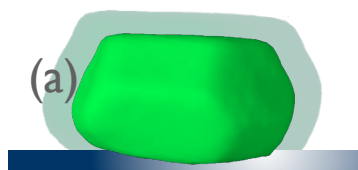


Reverse Problem Workflow

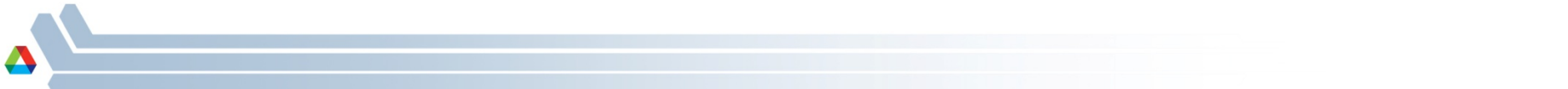


Case Study 3: Induced Shock Waves in Au Coated with Al

100 nm aluminum shell on 300 nm gold

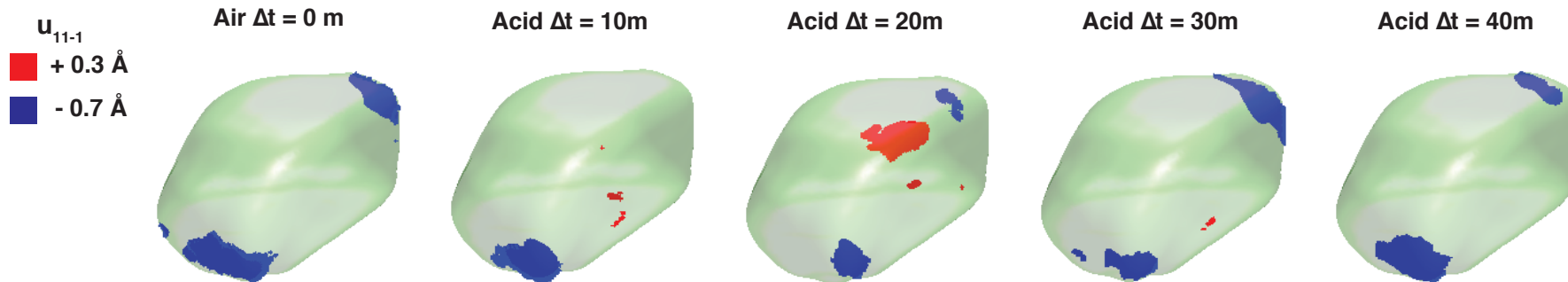


MD simulations → Calculated diffraction pattern

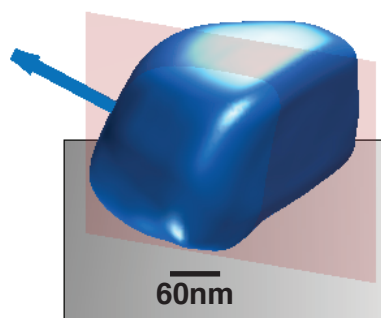


Case Study 4: Ascorbic Acid Decomposition on Gold

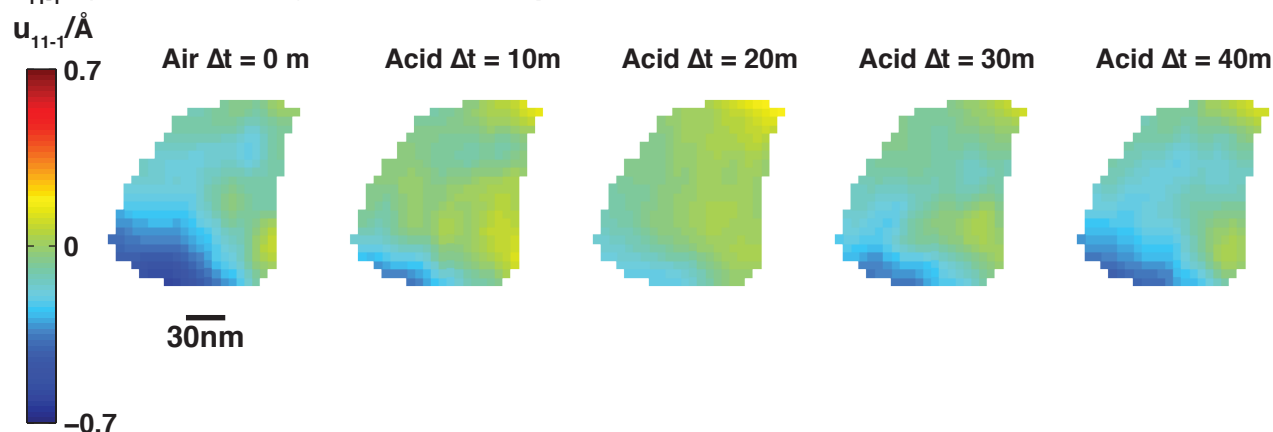
a) Isosurface of u_{11-1} values during first exposure



b) Cross-section Location



c) u_{11-1} dynamics during the first acid exposure

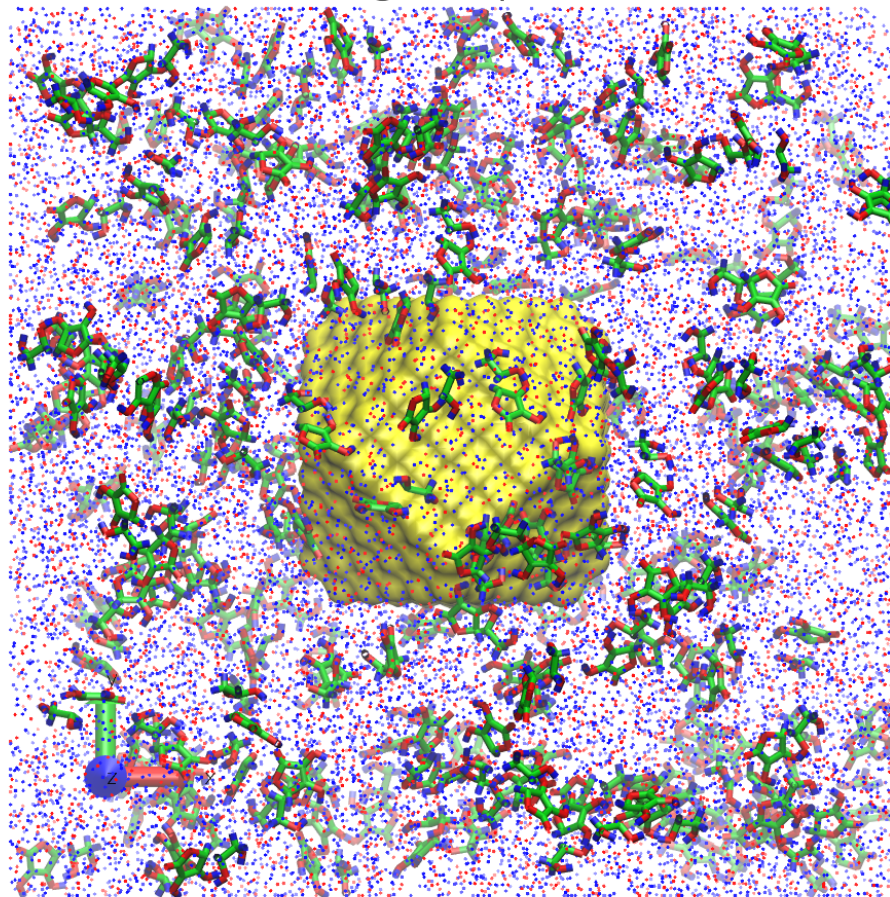


- Lattice change occurs at junctions involving the flattest facet
- Electron injection should create largest electric field at crystallographic discontinuities or apexes, providing “hot spots” for the reaction



Atomistic Simulation of Nanocatalytic Activity on Au Nanosurfaces

Sample schematic of simulated system
IM acid + water + gold (truncated octahedron)



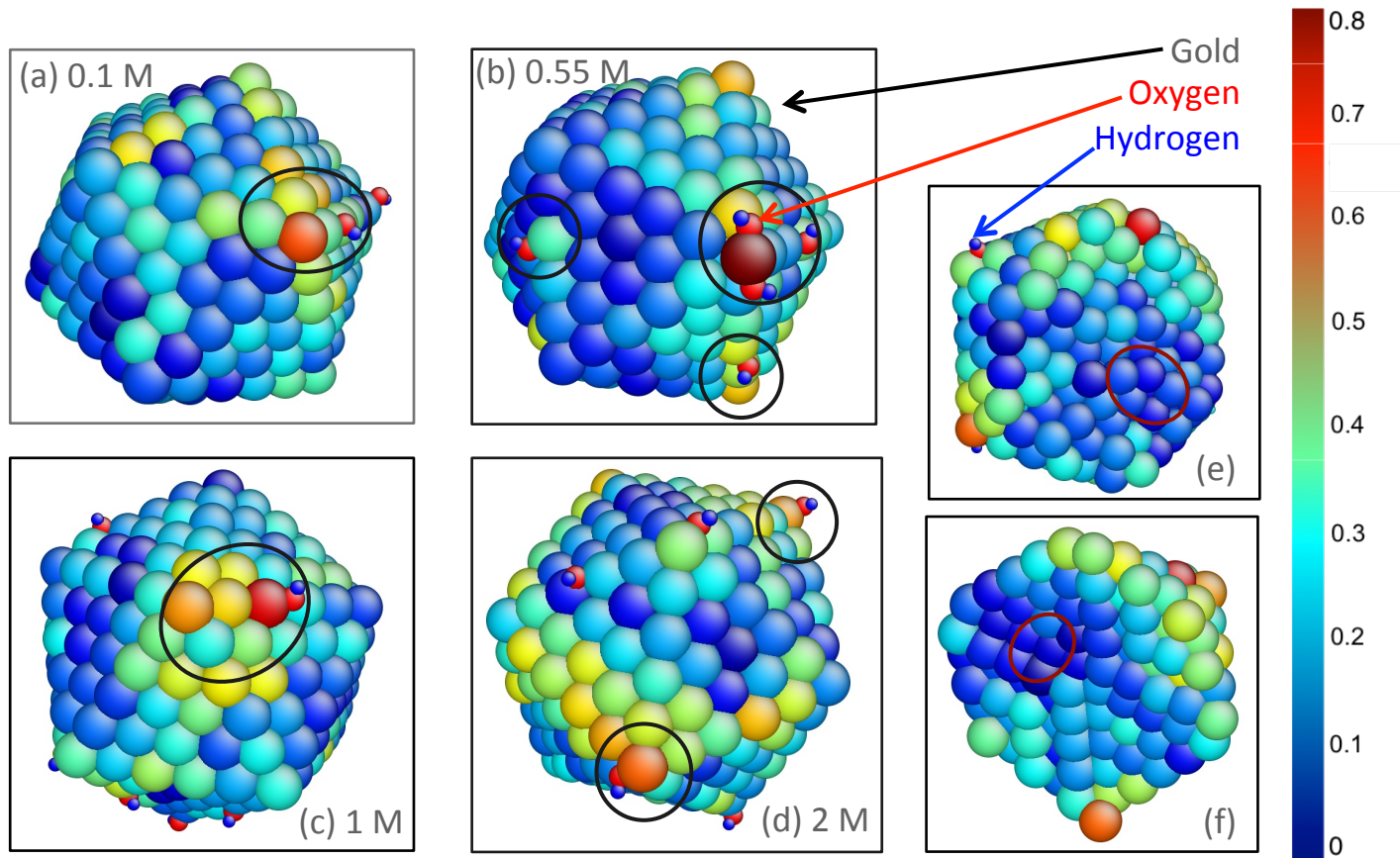
Yellow - Gold
Green - Carbon

Red - Oxygen
Blue - Hydrogen

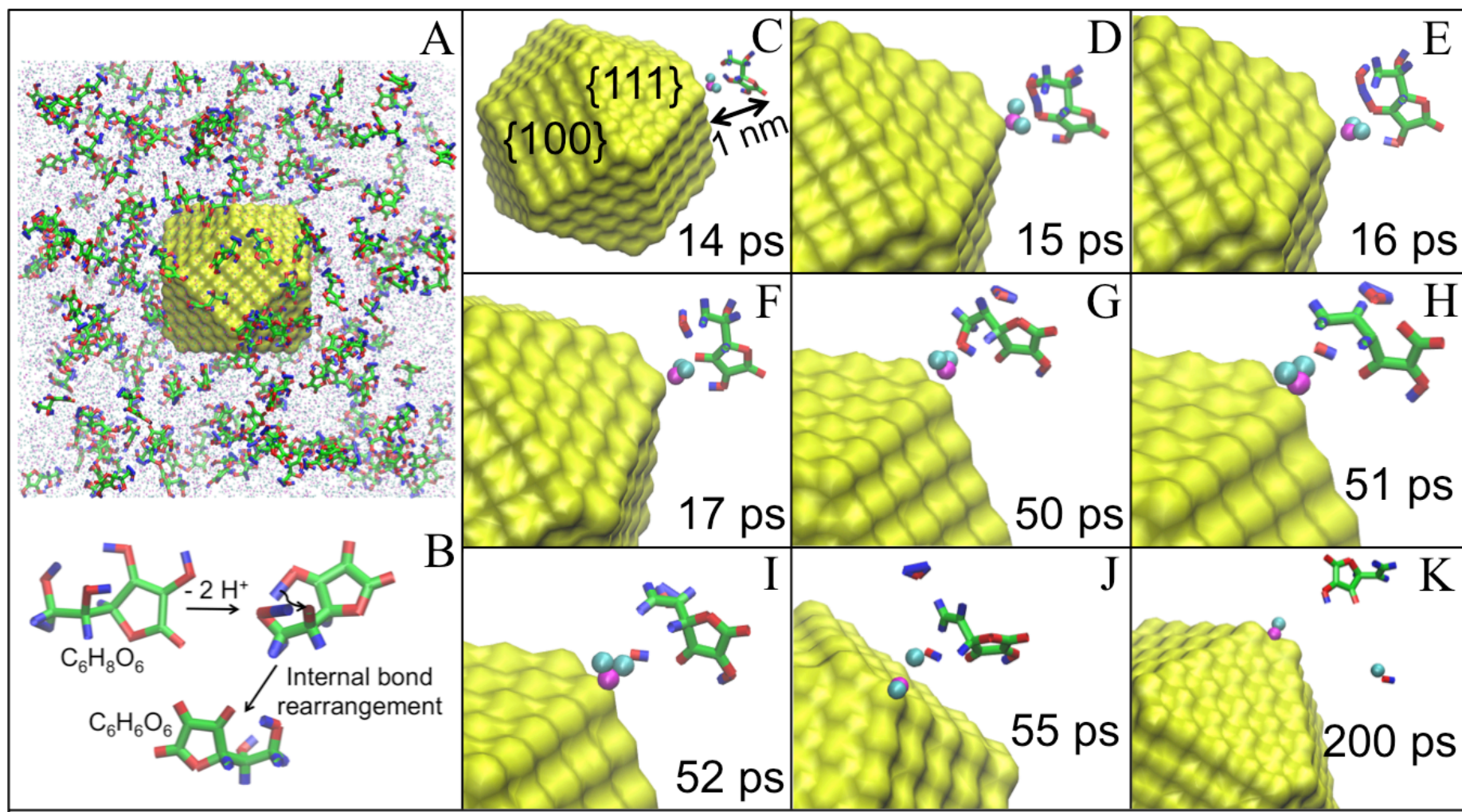


Effect of Acid Molarity

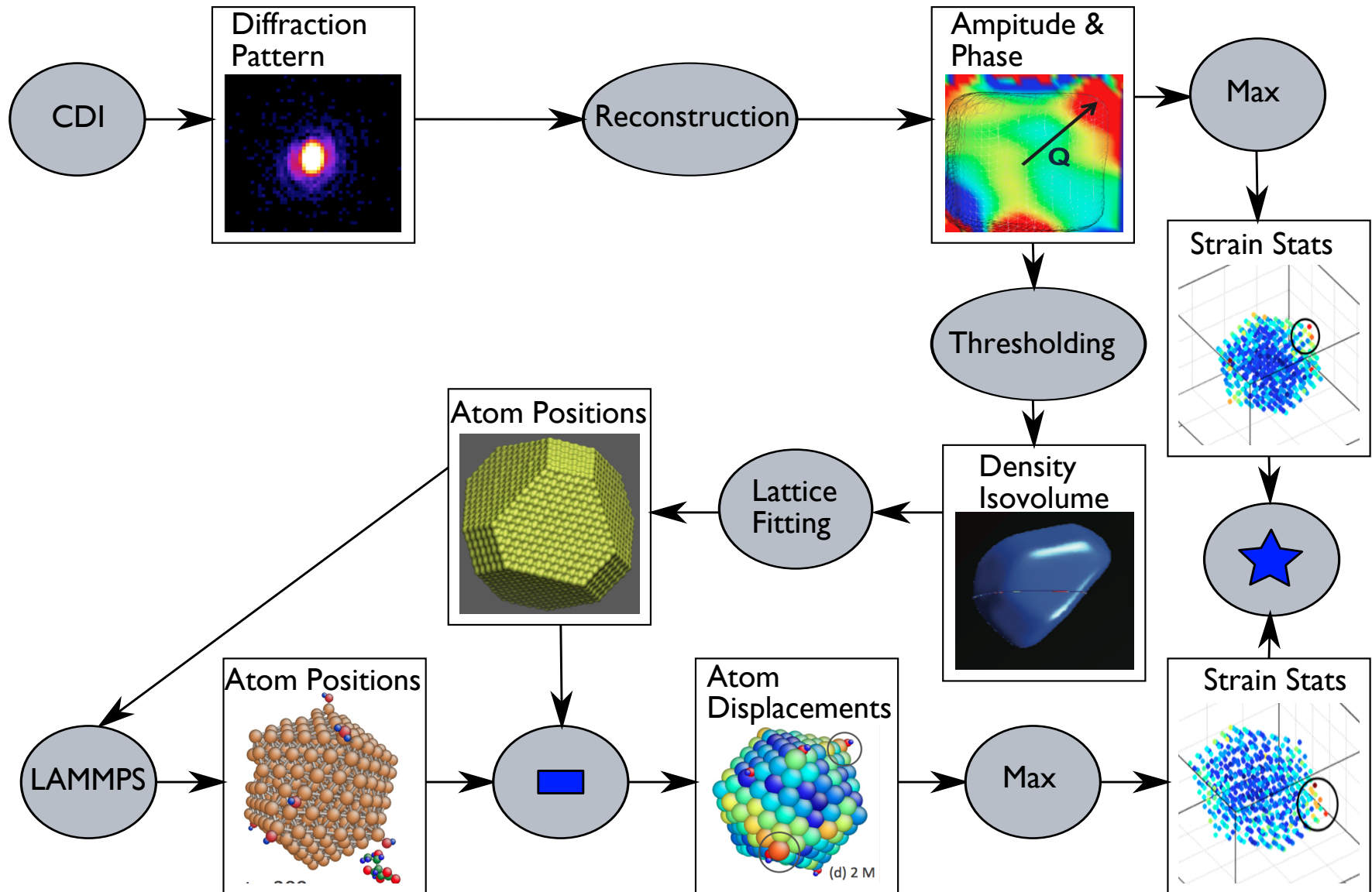
Displacement of Au atoms after 200 ps



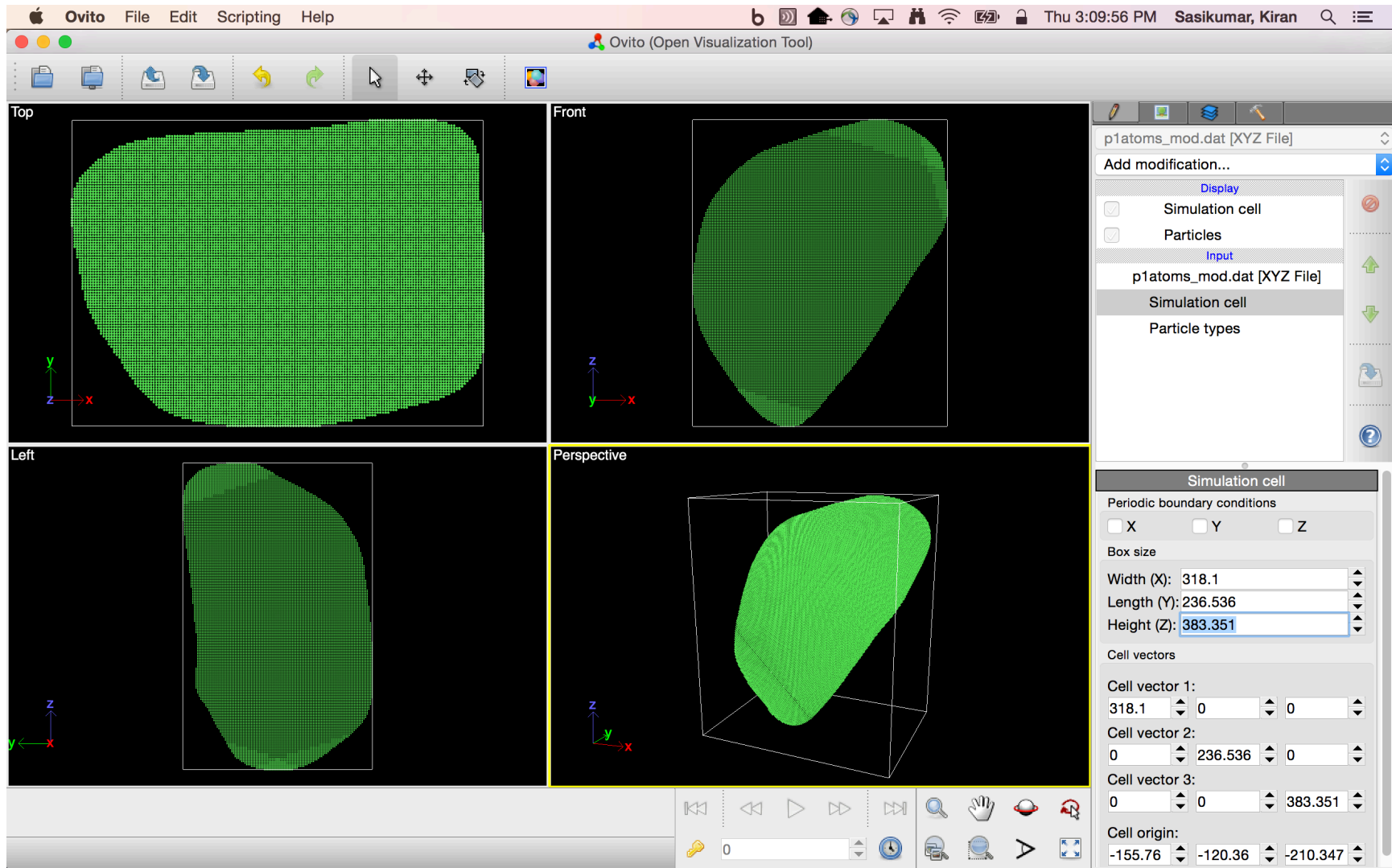
Adsorption Pathway



Forward Problem Workflow

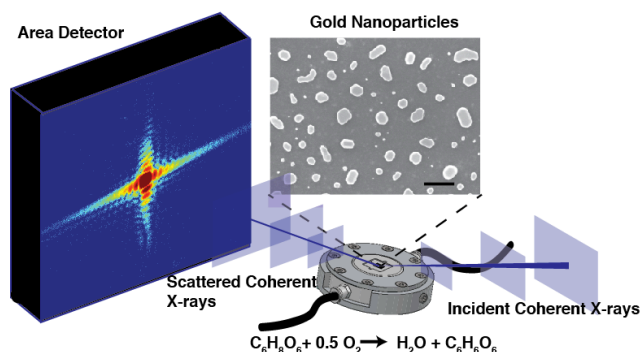


Lattice Fitting



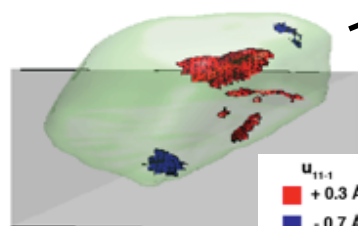
Modeling Ascorbic Acid Decomposition on Gold

CXDI Experiment



60nm

Acid $\Delta t = 20\text{ms}$



$u_{11-1}/\text{Å}$

0.7

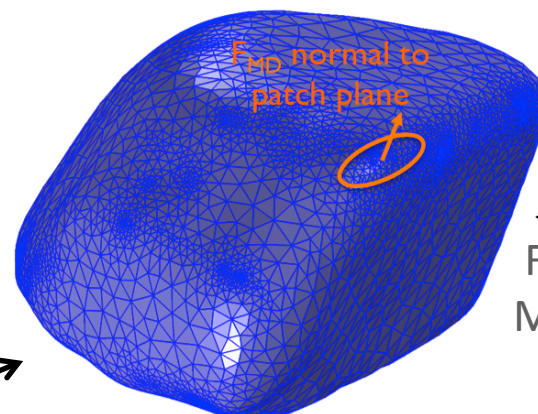
0

-0.7

30nm

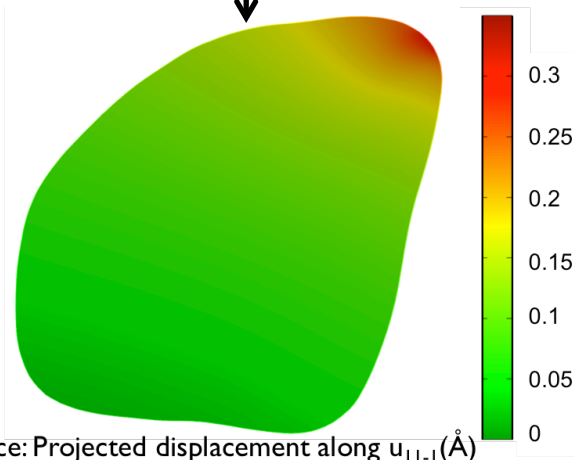
Experimental reconstruction as input to FEM

FEM (COMSOL)



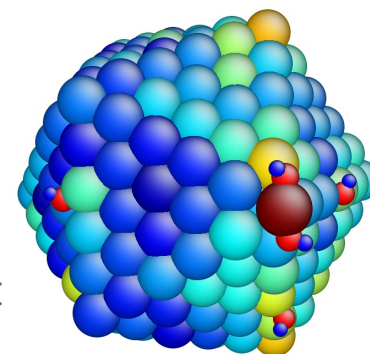
Minimum element size: 1 nm
Maximum element size: 20 nm

FEM Result



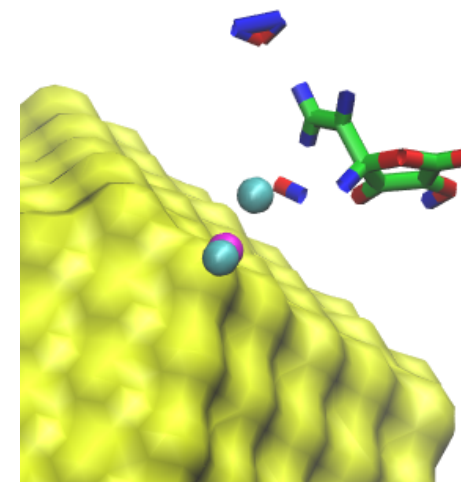
Experiment vs. Theory

MD (ReaxFF – LAMMPS)



Snapshot of Au and chemisorbed OH. Au colored by displacement from reference lattice.

Force from MD as input to FEM



MD also reveals underlying mechanism.

Food for Thought

■ Science challenges

- Time and space scales
 - Experiment: 10-20 nm resolution, particle size > 100 nm; time resolution 100 ps at the APS
 - Modeling: 1B atoms, 100 nm particle, 1 atom spatial resolution; time step 1 fs, 10s of ns duration
- Comparing experiment with model
 - Measure different quantities or the same quantity differently
 - We can measure strain on one lattice vector; how do we do that in 3d? In atomic scale?
 - Various techniques are appropriate for different scale models (eg FEM instead of MD)

■ CS challenges

- Language barriers
- Time and space scales
- Workflows: automation, reproducibility



Food for Thought

- Next steps
 - Draft paper for submission to a journal
 - Complete analysis of systems we measured ZnO and AuAl, MD model those systems
 - Corrosion experiment, submitted proposal for beam time at 34 and compute time Cu in salt water
 - In situ reconstruction and analysis
 - Machine learning to fit measured diffraction pattern to database of diffraction patterns (AICDI LDRD)



Funding Opportunities

- Workshops: Workflows workshop, Experimental and Observational Data workshop, Rumored upcoming experiment + simulation workshop
- ECP
- BESAC future directions
 - “Beyond Ideal Materials and Systems: Understanding the Critical Roles of Heterogeneity, Interfaces, and Disorder” (e.g., corrosion)



Workshop on the Future of Scientific Workflows

MissionThe mission of this workshop is to develop requirements for workflow methods and tools in a combined HPC and distributed work environment to enable science applications to better manage their end-to-end data flow. The following...



Data Management, Visualization, and Analysis of Experimental and Observational Data (EOD) Workshop

The Data Management, Visualization, and Analysis of Experimental and Observational Data (EOD) Workshop is aimed at helping ASCR and the research community better understand data management, analysis and visualization needs related to user...

**CHALLENGES AT THE
FRONTIERS OF MATTER
AND ENERGY:**
Transformative Opportunities
for Discovery Science

A Report from the Basic Energy Sciences Advisory Committee

References

- Maui website (public)
<http://tpeterka.github.io/maui-project/>
- Maui project site (private)
<https://bitbucket.org/tpeterka1/maui>



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